

Theory of Anomalous Hall Effect in a Heavy fermion System with a Strong Anisotropic Crystal Field

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In a heavy fermion system, there exists the anomalous Hall effect caused by localized f -orbital freedom, in addition to the normal Hall effect due to the Lorentz force. In 1994, we found that the Hall coefficient caused by the anomalous Hall effect (R_H^{AHE}) is predominant and the relation $R_H^{AHE} \propto \rho^2$ (ρ is the electrical resistivity) holds at low temperatures in many compounds. In this work, we study the system where the magnetic susceptibility is highly anisotropic due to the strong crystalline electric field on f -orbitals. Interestingly, we find that R_H^{AHE} is nearly isotropic in general. This tendency is frequently observed experimentally, which has casted suspicion that the anomalous Hall effect may be irrelevant in real materials. Our theory corresponds to corrections and generalizations of the pioneering work on ferromagnetic metals by Karplus and Luttinger.

KEYWORDS : heavy fermion system, anomalous Hall effect, periodic Anderson model, orbital degeneracy, CeRu₂Si₂

In many heavy fermion (HF) systems, the Hall coefficient (R_H) in a paramagnetic state shows universal temperature dependence, reflecting the crossover of the electronic property: As temperature increases, R_H also increases rapidly, and decreases after showing a peak at T_0 . [1–3] (we call T_0 the ‘coherent temperature’). The peak value $R_H(T_0)$ is apparently enhanced: it reaches $\sim 10^2$ times that of a normal metal. The origin of this interesting phenomena should be confirmed.

Today, some theoretical explanations have been proposed in terms of the anomalous Hall effect (AHE), which is caused by the f -orbital freedom. [4–6] In 1994, we proved the existence of the AHE in the periodic Anderson model (PAM) with orbital degeneracy, using the Kubo formula and the Fermi liquid theory. [4,7,8] The observed R_H in HF systems is given by $R_H = R_H^{AHE} + R_H^{\text{norm}}$, where R_H^{AHE} is the anomalous Hall coefficient we found and R_H^{norm} is the normal Hall coefficient caused by the Lorentz force, which is temperature independent below the coherent temperature. [9,10] The AHE naturally explains the sign and the magnitude of R_H in HF systems, and it shows a simple relation at low temperatures, $R_H^{AHE} = a \cdot \rho^2$, where ρ is the observed electrical resistivity. (see Fig. 11 of ref. [4].) The mechanism of our AHE is essentially similar to that found in ferromagnetic metal by Karplus and Luttinger. [4,11]

In this work, we extend this theory to an anisotropic

system with a crystalline electric field (CEF). Figure 1 shows anisotropy of R_H and χ in CeRu₂Si₂ [3], in which the relation $R_H \propto \rho^2$ holds at low temperatures, as shown by Fig. 2. While χ is extremely anisotropic, R_H is nearly isotropic. This appears quite paradoxical because AHE is the magnetic contribution and R_H^{AHE} is enhanced by χ in the spherical system [4]. We calculate the direction-dependence of both R_H^{AHE} and χ under the tetragonal CEF, and find that R_H^{AHE} is nearly isotropic even if χ is highly anisotropic, in general. This study confirms that AHE is predominant at low temperatures in many HF compounds.

The history of the study of AHE is very long, which started on the ferromagnetic metals. [11–13]. Here, we comment on the other theory on AHE in paramagnetic HF materials given by Fert and Levy, [6] which is the improvement of ref. [5]. They studied the impurity Anderson model ($J = 5/2$) with an additional scattering potential ($l = 2$). They calculated R_H^{AHE} using the Boltzmann equation for higher temperatures ($T > T_0$), where each f -electron site is almost incoherent. This mechanism is essentially the same as the theory of AHE in dirty ferromagnetic metal, given by Smit. [12] They concluded that $R_H^{AHE} \propto \chi \cdot \rho$ for $T > T_0$. Here we comment that their theory on AHE are essentially different from ours, which cannot be derived by the Boltzmann equation. For example, the simple extrapolation of our theory predicts that $R_H^{AHE} \propto \chi$ in the incoherent region ($T > T_0$), which is independent of ρ . Both of them may co-exist. But, it is definite that our AHE predominates at least for $T < T_0$.

Here, we study Ce-compounds, where f^1 configuration is realized. Due to strong l - s coupling, we have only to treat $J = 5/2$ sector, which has sixfold degeneracy. In a tetragonal crystal, this degeneracy splits into three levels by CEF, $|\Gamma\rangle = |\pm 1\rangle, |\pm 2\rangle, |\pm 3\rangle$, where $|+\Gamma\rangle$ and $|-\Gamma\rangle$ form a Kramers doublet. (Now, J_z is not a good quantum number.) In this work, we discuss the orbitally degenerate ($J = 5/2$) PAM under a tetragonal CEF as follows:

$$H = \sum_{\mathbf{k}\sigma} \epsilon_{\mathbf{k}} c_{\mathbf{k}\sigma}^\dagger c_{\mathbf{k}\sigma} + \sum_{\mathbf{k}\Gamma\Gamma'} E_{\Gamma\Gamma'}^f f_{\mathbf{k}\Gamma}^\dagger f_{\mathbf{k}\Gamma'} + \sum_{\mathbf{k}\Gamma\sigma} (V_{\mathbf{k}\Gamma\sigma}^* f_{\mathbf{k}\Gamma}^\dagger c_{\mathbf{k}\sigma} \\ + \text{h.c.}) + \frac{U}{2} \sum_{\mathbf{k}\mathbf{k}'\mathbf{q}\Gamma\Gamma'} f_{\mathbf{k}+\mathbf{q}\Gamma}^\dagger f_{\mathbf{k}'-\mathbf{q}\Gamma'}^\dagger f_{\mathbf{k}'\Gamma'} f_{\mathbf{k}\Gamma}, \quad (1)$$

where $c_{\mathbf{k}\sigma}^\dagger$ ($f_{\mathbf{k}\Gamma}^\dagger$) is the creation operator of the conduction electron (f -electron), and $\epsilon_{\mathbf{k}}$ ($E_{\Gamma\Gamma'}^f$) is the spectrum for conduction electrons (f -electrons). In a tetrag-

onal crystal, the f -orbital eigenstate $|\Gamma\rangle$ is given by $|\Gamma\rangle = \sum_M O_{\Gamma M} |J=5/2, J_z=M\rangle$, where $O_{\Gamma M}$ is an 6×6 orthogonal matrix and is given by

$$\begin{aligned} O_{\pm 1, \pm 5/2} &= O_{\pm 3, \pm 3/2} = a, \\ -O_{\pm 1, \mp 3/2} &= O_{\pm 3, \mp 5/2} = \sqrt{1-a^2}, \\ O_{\pm 2, \pm 1/2} &= 1, \end{aligned} \quad (2)$$

and other elements are zero. In a cubic crystal, $a = \sqrt{5/6} = 0.913$. (In a hexagonal crystal, $a = 1$.) Then, we can easily verify that $V_{\mathbf{k}\Gamma\sigma} = \sum_M O_{\Gamma M} V_{\mathbf{k}M\sigma}$ and $J_{\Gamma\Gamma'}^\gamma = \sum_{MM'} O_{\Gamma M} O_{\Gamma' M'} J_{MM'}^\gamma$ ($\gamma = x, y, z$), J^γ being the angular momentum along γ . (Note that $V_{\mathbf{k}M\sigma} = V_0 \cdot \sqrt{4\pi/3} \sum_m -\sigma \sqrt{(7/2 - M\sigma)/7} \delta_{m,M-\sigma/2} Y_{l=3}^m(\theta_k, \varphi_k)$ is derived for a spherical case, where $Y_{l=3}^m(\theta_k, \varphi_k)$ is the spherical harmonic function. [14]) Finally, $E_{\Gamma\Gamma'}^f$ under the magnetic field \mathbf{H} is given by $E_{\Gamma\Gamma'}^f = E_\Gamma^f \cdot \delta_{\Gamma\Gamma'} + g\mu_B \sum_\gamma J_{\Gamma\Gamma'}^\gamma \cdot H^\gamma$ up to H^1 -order, where g is the Lande's g -factor ($g=6/7$ for $J=5/2$), and we put $\mu_B = 1$ hereafter. For the time being, we limit our study to the case of $U = 0$, or to the mean-field (Gutzwiller) treatment: $V_{\mathbf{k}\Gamma\sigma} \rightarrow \sqrt{z_\Gamma} \cdot V_{\mathbf{k}\Gamma\sigma}$ and $E_\Gamma^f \rightarrow E_\Gamma^{f*} \equiv z_\Gamma \cdot (E_\Gamma^f - \mu) + \mu$, where z_Γ is the renormalization constant. [15] The electronic structure of orbitally degenerate $J = 5/2$ PAM is shown in Fig. 3, where $E_{\mathbf{k}}$ is the quasiparticle spectrum for \mathbf{k} , given by the pole of Green functions.

In the mean-field approximation, χ is given by

$$\begin{aligned} \chi &= \chi_P + \chi_V, \\ \chi_P &= \sum_\sigma \sum_{\mathbf{k}} \left(\frac{\partial E_{\mathbf{k}\sigma}}{\partial H} \right)^2 \cdot \delta(\mu - E_{\mathbf{k}}), \\ \chi_V &= - \sum_\sigma \sum_{\mathbf{k}} \frac{\partial^2 E_{\mathbf{k}\sigma}}{\partial H^2} \cdot \theta(\mu - E_{\mathbf{k}}), \end{aligned} \quad (3)$$

where χ_P and χ_V are called Pauli susceptibility and Van-Vleck susceptibility, respectively. Both χ_P and χ_V take the positive value [14]. After a long calculation, we get the expressions in the case of $\mathbf{H} \parallel \gamma$ ($\gamma = x, y, z$):

$$\chi_P^\gamma = g^2 \int \frac{d\Omega_{\mathbf{k}}}{4\pi} \rho_{\mathbf{k}}^c(\mu) \sum_{\sigma\sigma'} |M_{\mathbf{k}\sigma\sigma'}^\gamma(\mu)|^2, \quad (4)$$

$$\begin{aligned} \chi_V^\gamma &= -2g^2 \int \frac{d\Omega_{\mathbf{k}}}{4\pi} \int_{-D}^\mu \rho_{\mathbf{k}}^c(E_{\mathbf{k}}) dE_{\mathbf{k}} \\ &\times \left\{ \frac{\partial}{\partial E_{\mathbf{k}}} \left[\frac{1}{2} a_{\mathbf{k}}(E_{\mathbf{k}}) \sum_{\sigma\sigma'} |M_{\mathbf{k}\sigma\sigma'}^\gamma(E_{\mathbf{k}})|^2 \right] \right. \\ &\left. + \sum_{\sigma, \Gamma\Gamma'\Gamma''} \alpha_{\mathbf{k}\Gamma\sigma}(E_{\mathbf{k}}) \cdot \frac{J_{\Gamma\Gamma'}^\gamma J_{\Gamma'\Gamma''}^\gamma}{E_{\mathbf{k}} - E_{\Gamma'}^f} \cdot \alpha_{\mathbf{k}\Gamma''\sigma}^*(E_{\mathbf{k}}) \right\}, \end{aligned} \quad (5)$$

where $\rho_{\mathbf{k}}^c(\epsilon)$ represents the DOS for conduction electrons, $-D$ is the bottom of $E_{\mathbf{k}}$, and

$$M_{\mathbf{k}\sigma\sigma'}^\gamma(\epsilon) = \sum_{\Gamma\Gamma'} \alpha_{\mathbf{k}\Gamma\sigma}(\epsilon) J_{\Gamma\Gamma'}^\gamma \alpha_{\mathbf{k}\Gamma'\sigma'}^*(\epsilon), \quad (6)$$

$$\alpha_{\mathbf{k}\Gamma\sigma}(\epsilon) = V_{\mathbf{k}\Gamma\sigma}/(\epsilon - E_\Gamma^f), \quad (7)$$

$$a_{\mathbf{k}}(\epsilon) = \left(1 + \sum_{\Gamma} |\alpha_{\mathbf{k}\Gamma\sigma}(\epsilon)|^2 \right)^{-1}. \quad (8)$$

Note that $dE_{\mathbf{k}}/d\epsilon_{\mathbf{k}} = a_{\mathbf{k}}(E_{\mathbf{k}})$. The $E_{\mathbf{k}}$ -integration in (5) can be done easily when $\rho_{\mathbf{k}}^c(\epsilon)$ is constant. In the case of no CEF ($E_\Gamma^f = E^f$ for any Γ), the relation $\chi_P \approx \chi_V$ is derived by (4) and (5). In contrast, in the strong CEF case ($E_\Gamma^f \ll E_\Gamma^f, E_k^f$), χ_P shows strong anisotropy and $\chi_P \gg \chi_V$ is realized within the mean-field approximation. In reality, it is not easy to investigate the accuracy of the above mean-field results. In the case of no CEF, this problem is studied by the Fermi liquid theory, [16] or by the second-order perturbation theory w.r.t. U , [17] and the relation $\chi_P \approx \chi_V$ has been confirmed now.

On the other hand, the Hall conductivity due to AHE is given by eq. (2.16) in Ref. [4], using the Kubo formula. While we take the effect of CEF into account, we neglect the vertex corrections for currents. Under the magnetic field $\mathbf{H} \parallel \gamma$ ($\gamma = x, y, z$), the anomalous Hall conductivity $\sigma_{\alpha\beta}^{\text{AHE}}$ ($\epsilon_{\alpha\beta\gamma}=1$) is given by

$$\begin{aligned} \sigma_{\alpha\beta}^{\text{AHE}}/H &= g \frac{e^2}{2} \sum_{\sigma} \sum_{\Gamma\Gamma'\Gamma''} \int \frac{d\Omega_{\mathbf{k}}}{4\pi} \cdot \rho_{\mathbf{k}}^c(\mu) \cdot \frac{1}{\Delta_{\mathbf{k}}^c} \\ &\times \left[v_{\mathbf{k}\Gamma}^\alpha \cdot \frac{V_{\mathbf{k}\Gamma\sigma}^* V_{\mathbf{k}\Gamma'\sigma}}{(\mu - E_\Gamma^f + i\Delta_\Gamma)(\mu - E_{\Gamma'}^f - i\Delta_{\Gamma'})} \right. \\ &\cdot \left(\frac{v_{\mathbf{k}\Gamma'\Gamma''}^\beta J_{\Gamma''\Gamma}^\gamma}{\mu - E_{\Gamma''}^f + i\Delta_{\Gamma''}} + \frac{J_{\Gamma'\Gamma''}^\gamma v_{\mathbf{k}\Gamma''\Gamma'}^\beta}{\mu - E_{\Gamma''}^f - i\Delta_{\Gamma''}} \right) \\ &+ v_{\mathbf{k}\Gamma}^\beta \cdot \frac{V_{\mathbf{k}\Gamma\sigma}^* V_{\mathbf{k}\Gamma'\sigma}}{(\mu - E_\Gamma^f - i\Delta_\Gamma)(\mu - E_{\Gamma'}^f + i\Delta_{\Gamma'})} \\ &\left. \cdot \left(\frac{v_{\mathbf{k}\Gamma'\Gamma''}^\alpha J_{\Gamma''\Gamma}^\gamma}{\mu - E_{\Gamma''}^f - i\Delta_{\Gamma''}} + \frac{J_{\Gamma'\Gamma''}^\gamma v_{\mathbf{k}\Gamma''\Gamma'}^\alpha}{\mu - E_{\Gamma''}^f + i\Delta_{\Gamma''}} \right) \right], \end{aligned} \quad (9)$$

where

$$v_{\mathbf{k}\Gamma\Gamma'}^\alpha = \frac{\partial}{\partial k_\alpha} \left(\frac{\sum_{\sigma} V_{\mathbf{k}\Gamma\sigma}^* V_{\mathbf{k}\Gamma'\sigma}}{\mu - \epsilon_{\mathbf{k}}} \right), \quad (10)$$

$$v_{\mathbf{k}\Gamma}^\alpha = \sum_{\Gamma\Gamma'} \alpha_{\mathbf{k}\Gamma\sigma}(\mu) \alpha_{\mathbf{k}\Gamma'\sigma}^*(\mu) \cdot v_{\mathbf{k}\Gamma\Gamma'}^\alpha. \quad (11)$$

The derivative of the numerator in (10) gives the anomalous transverse current. In (9), Δ_Γ ($\Delta_{\mathbf{k}}^c$) is the damping rate of the f -electrons (conduction electrons), given by

$$\Delta_\Gamma = -\text{Im}\Sigma_\Gamma(\mu + i0) \quad (12)$$

$$\Delta_{\mathbf{k}}^c = \sum_{\Gamma} |\alpha_{\mathbf{k}\Gamma\sigma}(\mu)|^2 \cdot \Delta_\Gamma, \quad (13)$$

where $\Sigma_\Gamma(\omega)$ is the self-energy of f -electrons with Γ , which is \mathbf{k} -independent within the local approximation. Here, $\Sigma_\Gamma(\omega)$ is brought by the Coulomb interaction U . Within the second order perturbation and local approximation, taking the Pauli principle for Γ into account, it is easy to show that

$$\Delta_\Gamma = U^2 \cdot \frac{\pi}{2} \rho_\Gamma^f(\mu) \sum_{\Gamma' \neq \Gamma} \left(\rho_{\Gamma'}^f(\mu) \right)^2 \cdot (\pi T)^2 \quad (14)$$

for any Γ at low temperatures ($T \ll T_0$), where $\rho_\Gamma^f(\mu) \equiv \sum_{\mathbf{k}\sigma} |\alpha_{\mathbf{k}\Gamma\sigma}(\mu)|^2 \cdot \rho_{\mathbf{k}}^c(\mu)$ is the density of states for f -electrons with Γ at the Fermi energy. The relation (14) tells that $\Delta_\Gamma/\Delta_{\Gamma'}$ is temperature-independent for any Γ and Γ' at low temperatures, which will also be expected in the coherent region ($T \lesssim T_0$). As a result, $\sigma_{\alpha\beta}^{\text{AHE}}$ given by (9) is temperature-independent in the coherent region, where χ remains nearly constant and $\Delta_\Gamma \ll (E_\Gamma^f - \mu)$ is realized. [4] The definition of the Hall coefficient for $\mathbf{H} \parallel \gamma$ is $R_H = (\sigma_{\alpha\beta}/H)/(\sigma_{\alpha\alpha}\sigma_{\beta\beta})$ (where $\epsilon_{\alpha\beta\gamma}=1$). Thus, the relation $R_H^{\text{AHE}} \propto \rho^2$ is confirmed, as shown in Fig. 2.

The theory of Karplus and Luttinger contains a serious inconsistency, pointed out by ref. [12]. In ref. [4], we solved it by taking the dissipation of quasiparticles into account. The new aspects of the AHE we find are as follows: (i) We derived the exact form of $\sigma_{\alpha\beta}^{\text{AHE}}$, and found that it depends on the ratio $\Delta_\Gamma/\Delta_{\Gamma'}$, which differs greatly from 1 in the anisotropic system. This means that $\sigma_{\alpha\beta}^{\text{AHE}}$ depends on the origin of the quasi-particle damping. (ii) In our theory, the orbital moment ($g\hat{J}^\gamma$) plays an important role, and the l - s coupling term is not essential.

Following are the results of our numerical calculations on χ and $\sigma_{\alpha\beta}^{\text{AHE}}/H$, given respectively by (3) and (9). Now we put $z_\Gamma = 1$. Here, we study the case $\Delta_{1-2}, \Delta_{2-3} > 0$, where $\Delta_{1-2} = E_2^f - E_1^f$ and $\Delta_{2-3} = E_3^f - E_2^f$. Taking the experimental data on CeRu₂Si₂ into account, we put $\Delta_{2-3} = 3 \cdot \Delta_{1-2}$ [21]. For simplicity, we treat Δ_{1-2} and the parameter a independently. Other parameters we use are $E_1^f = -0.2$, $\mu = -0.23$, and $V_0 = 0.2$. We also put $\epsilon_{\mathbf{k}} = -1 + 2 \cdot (\mathbf{k}/\pi)^3$, where $\rho_{\mathbf{k}}^c = k^2 \cdot d\epsilon_{\mathbf{k}}/dk$ is \mathbf{k} -independent. These selections (and the shape of the Fermi surface) is to be irrelevant to the conclusion qualitatively. Figure 4 shows the a -dependence of χ^z/χ^x and $\sigma_{xy}^{\text{AHE}}/\sigma_{yz}^{\text{AHE}}$ in the case of $\Delta_{1-2} = 0.3$. We can recognize the isotropic feature of the anomalous Hall coefficient, which is quantitatively affected by the form of the damping rate. (When $\Delta_{1-2} = \Delta_{2-3} = 0$, both χ and σ_{ab} are isotropic and independent of a .) Figure 5 shows the dependence of χ^z , χ^x , σ_{xy}^{AHE} and σ_{yz}^{AHE} on Δ_{1-2} , in the case of $a = 0.96$. We can see that R_H is less affected by Δ_{1-2} than χ . As a result, R_H^{AHE} can take a large value even in strongly anisotropic systems. Especially, R_H^{AHE} tends to be isotropic for larger Δ_{1-2} ($\Delta_{1-2} \gg |E_1^f - \mu|$).

Such a difference of the anisotropy between on χ and on $\sigma_{\alpha\beta}^{\text{AHE}}$ can be understood naturally as follows: χ_P , given by (4), comes mainly from the lowest Kramers doublet (say, $\Gamma = \pm 1$). On the contrary, higher Kramers doublets are indispensable for $\sigma_{\alpha\beta}^{\text{AHE}}$, given by (9), because AHE is the multi-band contribution. [4] Thus, $\sigma_{\alpha\beta}^{\text{AHE}}$ is averaged and is nearly independent of directions. In conclusion, we find that the anisotropies of χ and R_H^{AHE} can be quite different. In the strongly anisotropic system due to the

(tetragonal) CEF, in general, χ is strongly anisotropic and R_H^{AHE} tends to be isotropic.

Finally, we point out some remaining problems. The present model may be too simplified for quantitative study in that it contains only one spherical conduction band $\epsilon_{\mathbf{k}}$. Besides, some kinds of vertex corrections (or $1/z_\Gamma$) omitted in this work should be studied for complete analysis. Moreover, the consideration on the Umklapp processes will be necessary for some compounds: References [4,8] show that $\sigma_{xx} \propto \Delta_c^{-1} \cdot C_{\text{UM}}$ and $\sigma_{xy}^{\text{AHE}}/H \propto \{C_{\text{UM}}\}^2$, thus $R_H \propto \Delta_c^2 \cdot \{C_{\text{UM}}\}^0$, where the coefficient C_{UM} reflects the Umklapp processes for electron-electron scattering, which dominate the various transport phenomena at low temperatures. [18] (Note that the Umklapp processes are irrelevant to R_H .) $C_{\text{UM}} \sim O(1)$ is satisfied in the usual case. But $C_{\text{UM}} \sim 10$ will be realized in CeRu₂Si₂, which shows large $\sigma_{xy}^{\text{AHE}}/H$ at low temperatures. (see Fig. 2 or ref. [4].) At last, we comment on the existence of the AHE in other kinds of metals. For example, at low temperatures, R_H in Sr₂RuO₄ shows the temperature dependence similar to that in HF compounds. [19] We find that the relation $R_H \propto \rho^2$ holds for $T \lesssim 6\text{K}$. [20] We believe that the AHE discussed in this work is general to many non-magnetic metals with orbital-freedom.

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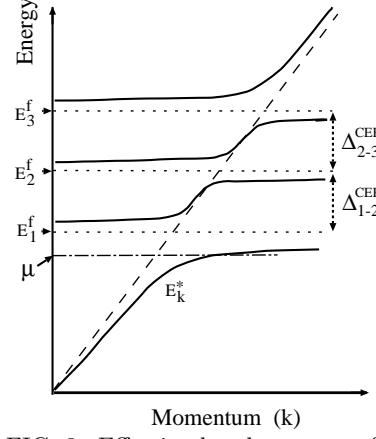


FIG. 3. Effective band structure for $J = 5/2$ PAM under the tetragonal CEF. μ is the Fermi energy.

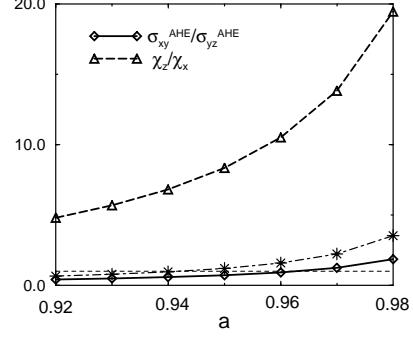


FIG. 4. a -dependence of the anisotropy of χ and σ^{AHE} . (a characterizes the CEF introduced by eq. (2).) Here, we set $\Delta_{1-2} = 0.3$ and $|E_1^f - \mu| = 0.03$. In comparison, we also show $\sigma_{yz}^{\text{AHE}}/\sigma_{xy}^{\text{AHE}}$ for the case of $\Delta_\Gamma = \text{constant}$, instead of eq. (14), by stars (dash-dotted line).

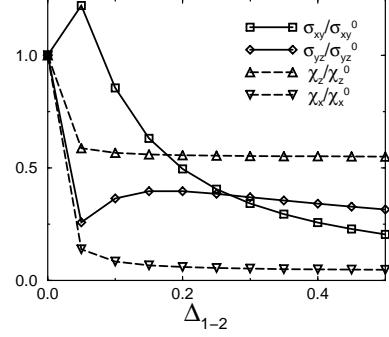


FIG. 5. Δ_{CEF} -dependence of χ^z , χ^x , σ_{xy}^{AHE} ($\mathbf{H} \parallel \mathbf{z}$) and σ_{yz}^{AHE} ($\mathbf{H} \parallel \mathbf{x}$). Here, we set $a = 0.96$ and $|E_1^f - \mu| = 0.03$.

FIG. 2. R_H vs ρ^2 for CeRu₂Si₂. This figure is borrowed from ref. [3].